THEORETICAL MODELING AND EXPERIMENTAL CHARACTERIZATION OF FIBER-MATRIX INTERFACE IN ADVANCED COMPOSITES

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INTRODUCTION

In this paper, a recently developed revolutionary approach to material characterization for application to the development of advanced composite materials is presented. A methodology of fiber-matrix interface characterization has been recently developed based on the shear back reflectivity technique. This new methodology uses ultrasonic waves to nondestructively evaluate the interfacial elastic properties at a localized level. This new approach is nondestructive in nature in contrast to traditional destructive characterization techniques which are costly, time consuming, tedious and which also render the samples unusable for other experiments. Based on this interface characterization methodology, an original, unique, innovative method of imaging fiber microcracking has been developed to augment standard experiments such as fiber fragmentation, transverse loading, fiber-push tests, etc., conducted by scientists developing advanced composites. This provides critical information to materials researchers for micro-mechanics modeling studies to evaluate fracture and fatigue behavior of composites. Further, the methodology is used to assist and enhance the traditional experiments such as fiber fragmentation tests for interfacial shear load transfer behavior studies, fiber push tests for interfacial frictional and fracture behavior studies, transverse loading tests for interfacial stress at fracture, etc. Thus, the nondestructive ultrasonic technique has been shown here to extend the horizons of material behavior studies by providing new information obtained during in situ testing or in an interrupted mode. Moreover, the new technique alleviate the need for traditional destructive characterization work such as optical metallography. The entirely new information provided by this breakthrough technique is now extensively used by materials behavior groups at the WL / Materials Directorate (WL/ML) for determination of the basic mechanisms of damage in composites to improve both the processing and the reliability of the experimental characterization of the materials.

BACKGROUND INFORMATION

An ultrasonic shear back-reflectivity technique (SBR) has been developed [1] to complement other existing techniques for the characterization of the interfacial behavior in fiber reinforced composites. The ultrasonic characterization of the interface is achieved by the analysis of the back-reflected signal from the fiber-matrix interface.

Theory

For the development of the theoretical model, consider a plane elastic wave propagating in the positive direction z_i, and obliquely incident at an angle θ on a model

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composite immersed in water, made with a cylindrical isotropic and homogeneous fiber embedded in a ceramic matrix. The shear properties of the matrix/fiber interface are modeled by assuming a shear, weightless, and inertia free spring [1] between the matrix and the fiber with an equivalent rigidity of N_S [GPa/μm]. The parameter N_S which represents the various interfacial conditions, is linked directly to the degree of discontinuity of the shear displacements at the interface. Hence, the shear stresses, σ^T, transmitted across the interface via the spring are given by the relationship:

$$\sigma^T = N_S \ [u^T] \qquad (1)$$

where $[u^T]$ denotes the jump of shear displacements across the interface. The linearity of equation (1) is based on the assumption of small amplitudes of vibrations which is justified in the case of ultrasonic elastic waves.

The shear back-reflection coefficient from the fiber is calculated by [1]:

$$SBR = T_M \ R_F \ T_W \qquad (2)$$

where T_M, T_W are transmission coefficients at the water/matrix and matrix/water boundaries and R_F is the reflection coefficient at the fiber-matrix interface. Further details can be found from the literature [1].

Experimental Procedure for Interface Elastic Property Measurement

The experimental procedure for the determination of the shear stiffness coefficient of the fiber/matrix interface consists of (a) measuring the shear wave back reflection coefficient of the interface and, (b) inverting the calculation of the shear stiffness coefficient by using the relationship between N_S and back-reflection coefficient established by using the theoretical model for an appropriate ultrasonic frequency of interrogation.

An experimental sample block was fabricated for the measurement of the reflection coefficient because the calculation of the reflection coefficient requires the measurement of the wave amplitude incident on the interface. Air was sealed in behind the angle surface wherein the machined angle is the same as the refracted shear wave angle. The sample also had a drilled hole with the same diameter as that of the fiber (142 microns). The drilled hole was sealed to keep out the coupling fluid (water) and was used as a target to simulate an unbonded interface. Finally, the sample also had a SCS-6 fiber embedded in the matrix material (either Ti-6Al-4V or Ti-24Al-11Nb). The experimental procedure for the measurement of the interfacial shear stiffness coefficient was carried out in several steps. First, a reference A-scan was obtained from the angled surface. A Fourier transformation provided the incident magnitude at 25 MHz. Similarly, A-scans were obtained from the hole and the fiber and the corresponding reflected magnitudes at 25 MHz were measured using Fourier transformation and the reflection coefficient was calculated. The calculated reflection coefficient was used to inverse calculate the shear stiffness coefficient of the equivalent elastic interface using the theoretical curve generated from the model described in the previous section. Further details of this experimental measurement process can be found in the literature [2].

INTERFACE EVALUATION OF MODEL COMPOSITES USING ULTRASONICALLY ENHANCED MECHANICAL DESTRUCTIVE TESTS

The characterization of the interface is of great interest to the researchers who are developing new composite systems. In order to determine the optimum interface properties, a model monofilament composite is usually used which has an interface with tailored properties depending on the choice of the fiber and the matrix materials, the fiber coating and the processing conditions. Many investigators have employed a number of

experimental destructive techniques including fiber pull-out, fiber push-out, fiber fragmentation, transverse loading tests, etc. to characterize the interface in model composites. This section presents results which illustrate the benefits of concurrent use of the above mentioned destructive techniques and the ultrasonic nondestructive characterization techniques that have been recently developed in the Materials Directorate for interface evaluation in various stages of development of advanced composite materials.

Influence of the Interface Elastic Properties on CMC Micro-Cracking

Recent analytical and experimental work [3-6] with glass-ceramic composites containing Nicalon fibers in a Calcium-Aluminosilicate (CAS) matrix has supplied evidence that the increase in crack growth resistance noted in regions of high fiber volume fraction may in turn translate into superior microcrack initiation stress levels for composites with uniformly and closely spaced fibers. In this work, the emphasis was on the initiation of microcracking under tensile loading of unidirectional SiC fiber reinforced, borosilicate glass matrix composites with controlled spacing and different fiber-matrix interface properties as well as residual stresses.

The glass was supplied by Corning as an ~8μm powder. The composition of the glass was varied so as to vary the thermal expansion coefficient mismatch between the fiber and the matrix. The resulting residual radial thermal stresses after processing could be varied from tensile to compressive. In addition, two different fibers have been utilized: a TiB_2 coated SIGMA fiber with a diameter of 102 μm and a carbon coated SCS-6 fiber with a diameter of 142 μm. The borosilicate glass strongly wets the TiB_2 coating of the SIGMA fiber while it weakly wets the carbon coating of the SCS-6 fiber. This allowed us to investigate the effects of bonding at the fiber-matrix interface as well as fiber diameter upon the initiation of matrix cracking. The composites were processed by tape casting the glass powder into a green tape with a relative density of 50%. The green tapes were cut to size and laminated with fiber mats of the desired fiber spacing (68 or 120 fibers per inch). The volume fraction of fibers in the composites was varied by altering the thickness of the green tape and using the two different fiber spacings. After lamination the composites were inserted into a tube furnace and vacuum sintered at 710° C for one hour. The samples were then hot isostatically pressed at 650° C for 30 min. with an applied pressure of 35 MPa to remove the residual porosity (~2%). The resulting samples were approximately 10 cm long by 2 cm wide with a thickness of 0.2 cm.

The composite cracking stress of the samples was evaluated using a procedure outlined in [3]. Fiber volume fractions were determined using the rule of mixtures with the moduli of the respective fibers and matrix which were determined by separate tests. Axial and transverse strains were measured using strain gages. Free edges and flat surfaces of specimens were polished with diamond paste in order to enhance microscopic imaging for crack detection. The upper and lower surfaces were coated with a thin layer of epoxy to reduce the dominance of surface flaws. Acoustic emission and photomicrographic techniques were employed for detection of initial matrix cracking. Micrographs were mainly used to verify the acoustic emission results through physical observation of cracks. Initial cracking was consistent with the full-cell mode [4] except in one specimen.

From the model predictions [5] for the SIGMA and SCS-6 fibers in a 7040 borosilicate ($\Delta\alpha$ = -1.4 μm/μm/°C, i.e. a compressive stress upon the fiber at the fiber/matrix interface) glass matrix, the effect of a residual thermal stress due to processing was to lower the level of stress at which initial matrix cracking occurs in the 7040 glass matrix. In addition, the lower the fiber diameter, the higher the matrix cracking stress. The experimental results for the SIGMA fibers in 7040 glass match the model prediction quite well while those of the SCS-6 fiber reinforced composites appear to suggest debonding of the fiber-matrix interface. Such a debonding was possible because, the borosilicate glasses do not strongly wet the carbon coating of the SCS-6 fiber thereby producing a weak interface.

Following testing, several specimens were ultrasonically evaluated to (1) assess the interfacial bonding, and (2) to determine the amount of microcracking for corroboration with the optical micrographs of surface cracks. While ultrasonic imaging and evaluation of the composites with SIGMA fibers did not show dramatic debonding and cracking behaviors, the composites with SCS-6 fibers indicated poor interfacial bond integrity. Acoustic microscopy of the sample was also performed wherein large areas of fiber/matrix debond were observed (Figure 1). This debonding behavior indicates a weak interfacial bonding compared to SIGMA fibers. Additional experiments were conducted to confirm this observation. Shear back reflectivity technique (SBR) was used [1] to quantify the stiffness coefficient of the fiber/matrix interface for both SIGMA and SCS-6 fibers embedded in 7040 Glass as well as Glass-E. The results are tabulated in Table 1 and were obtained using the theoretical curves from the model. The interface quantification process indicates that the interface for 7040-SIGMA is relatively stiff compared to the other interfaces with the Glass-e/SCS-6 interface being the most compliant. Further details of this work can be found in the literature [7].

Single-Fiber Fragmentation Test for Metal Matrix Composites

In the fiber fragmentation test, a composite sample made of a single fiber embedded in a ductile matrix is subjected to tensile loading along the fiber axis [8,9]. When the tensile stress, which is transferred from the matrix to the fiber by shear, exceeds the local fiber strength, the single fiber breaks successively into smaller fragments until the fragments become too short to enable further increase in stress level. Using arguments based on shear lag analysis, Kelly and Tyson [8] showed that the critical length of fiber for load transfer, L_c, is a function of the interfacial shear stress according to the equation

$$\tau_i = \sigma_f \frac{d}{2L_c}$$

where τ_i is the shear stress, σ_f is the tensile strength of the fiber of critical length, L_c, and d is the fiber diameter. As a result, the measurement of fiber fragment size is critical to the understanding of the load transfer behavior of the fiber-matrix interface. However, the metallographic approach to measure the fiber fragmentation size might induce further fragmentation. Also, even after a laborious and meticulous sample preparation for metallography, parts of the fiber can still retain remnants of the matrix and/or interphase materials thereby masking any fiber fracture present at that location. Therefore, a nondestructive method of imaging would improve the reliability of the measurement of the lengths of the fragments of the embedded fiber. In the next several sections, we present a revolutionary nondestructive ultrasonic approach developed [10] to image the fragmented fiber still embedded in the matrix. This is the first time ever that fiber fragmentation has been nondestructively imaged in opaque matrices.

The fiber fragmentation behavior of SIGMA SiC or SCS-6 SiC fiber reinforced Ti-6Al-4V and Ti-14Al-21Nb (both compositions in wt. %) was studied in order to determine the role of fiber/matrix interface characteristics in load transfer. The composite samples were fabricated by diffusion bonding (vacuum hot pressing at 925°C/5.5 MPa/30min followed by hot isostatic pressing at 1010°C/100 MPa/2 hr) two matrix alloy sheets with a single fiber between them. The consolidated samples were machined into 1.5 mm thick sheet tensile specimens with 19.05 mm x 6.35 mm gage sections. The tested specimens were ultrasonically imaged, sectioned and polished parallel to the fiber axis. Metallographic examination of the fiber fragments was conducted by using optical microscopy and SEM.

Figure 2a shows the image of a SCS-6 fiber embedded in Titanium Aluminide matrix (Ti-6Al-4V) and before any loading. Figure 2b shows the fiber after loading. Since the average fragment size is about the same as the diameter of the fiber, the interface has

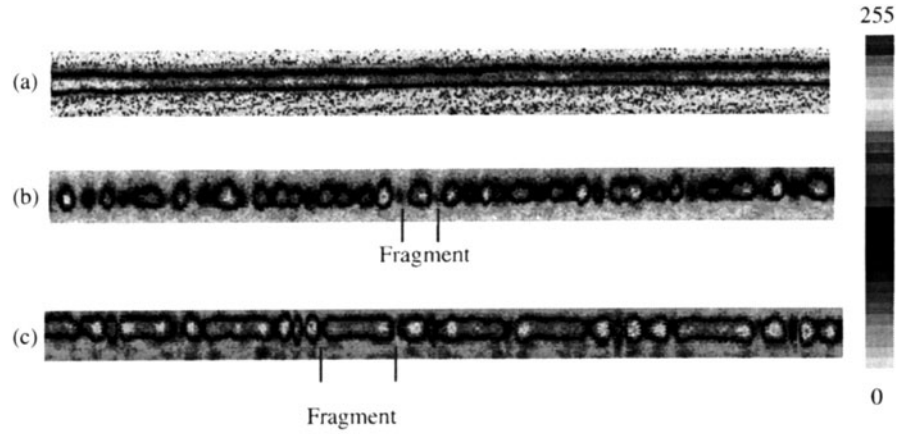
Figure 1 Acoustic Microscopy Image Showing Surface Cracks and Interfacial Debonding of the first layer of fibers.

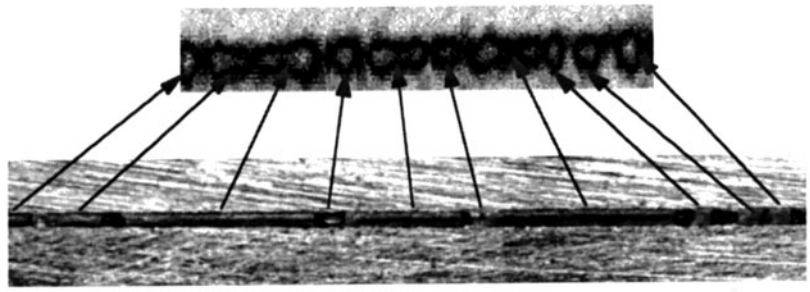
successfully transferred the load to the fiber. Figure 2c shows another sample with SCS-6 fiber embedded in Ti-14Al-21Nb matrix after loading. Since the average fragment size is more than three times the diameter of the fiber, the interface has less efficiently transferred the load to the fiber compared to the sample shown in Figure 2b. In both the 'after-test' images, in addition to the main breaks, smaller pieces due to secondary breaks are also observed in both Figures 2b and 2c. The presence of such secondary pieces have been corroborated by metallography (Figure 2d and 2e).

Single fiber fragmentation tests with continuous silicon carbide fibers in a Ti-6Al-4V alloy matrix were conducted with *in situ* ultrasonic imaging to monitor the fragmentation process. Straining proceeded incrementally on a specially designed load frame with acoustic emission detection (AE) performed during each increment and shear wave back-reflectivity (SBR) ultrasound images acquired following each increment. Metallographic examination of the fragmented fiber was performed following the straining sequence by electropolishing and scanning electron microscopy. A single fiber fragmentation specimen imaged by reflected ultrasound is shown in Figure 3, with increasing coupon strain levels moving from top to bottom in nominal strain increments of 0.75%. Ultrasonically detected breaks using SBR technique were compared by SEM performed after the final loading of the sample. We found that the SBR ultrasound image indicated the presence of a fiber break that was not visible under the SEM, but was apparently concealed by the reaction zone, which is not removed by electropolishing and was masking these region of the fiber. This interpretation was supported by the identification of other previously concealed fiber breaks by additional polishing. Changes in the polishing solution to allow it to attack the reaction zone and completely expose the fiber are under investigation. Before fragmentation begins, the fiber/matrix interface

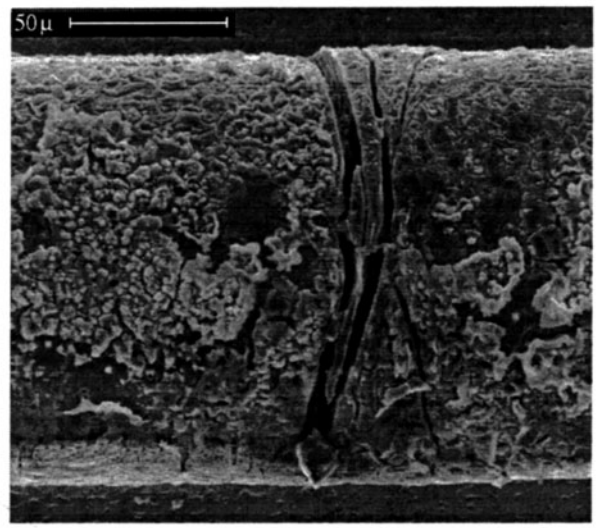
Table 1 Calculated values of the shear stiffness coefficients obtained from experimentally measured back-reflection coefficients for four different composite systems.

Composite system	Reflection Coefficient	Shear Stiffness Coefficient (GPa/μm)
7040/SIGMA	0.242	17
Glass-E/SIGMA	0.231	2.8
7040/SCS-6	0.318	3.2
Glass-E/SCS-6	0.208	1.3





(d)



(e)

Figure 2 (a) Ultrasonic Image of an untested single fiber sample. (b) Fiber Fragmentation Image in Ti-6Al-4V sample with SCS-6 Fiber. (c) Fiber Fragmentation Image in Ti-14Al-21Nb (or Ti-24Al-11Nb by atomic %) sample with SCS-6 Fiber. (d) Corroboration of the Ultrasonic Imaging of Fiber Fragmentation with Metallography. (e) SEM image of the fragmented fiber showing secondary fractures in addition to the primary fracture.

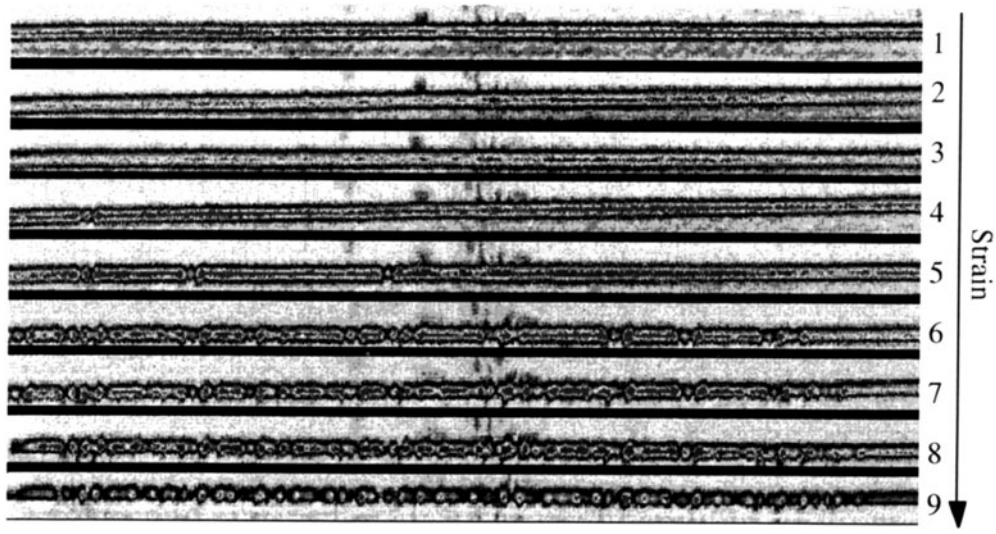


Figure 3 In-Situ Ultrasonic Images of the Fiber Fragmentation Process. Images 1 Through 9 are Sequentially Obtained by Imaging the Same Fiber Through the Loading Process with the Strain Increasing at a Nominal Rate of 0.75% from 1 Through 9.

uniformly reflects the ultrasound, producing the homogeneous band in the topmost image. The first fiber breaks produce clear patterns consisting of pairs of dark regions where the sound reflecting from the new surfaces created by the fiber fracture destructively interferes, with a central bright region where constructive interference occurs [10]. When the spacing between fiber breaks is large the pattern of constructive and destructive interference is unambiguous and easy to interpret in terms of break locations. With smaller break-to-break spacing at the end of the test, however, the observed ultrasound interference patterns may be the result of more than one possible fiber break arrangement and interpretation becomes more difficult. Further details of this work can be found in the literature [11].

Transverse Loading Test for Metal Matrix Composites

An understanding of the dependence of the fiber-matrix interface deformation and debonding on residual stresses, the fiber-matrix bond strength, and matrix properties under transverse loading conditions is needed for the improvement of the transverse properties of titanium matrix composites (TMC) reinforced with continuous silicon carbide (SiC) fibers. A new methodology has been developed to assess the interfacial stress at fracture. The newly developed method is based on an ultrasonic NDE technique which is used in-situ to monitor the deformation and failure of the fiber-matrix interface under transverse loading. Details of this work can be found in a paper published in this proceeding [12].

SUMMARY

This paper outlines the development, validation, and a successful technology transfer of an ultrasonic nondestructive method of fiber-matrix interface characterization. Summaries of the theoretical development, experimental quantification, and various applications of the techniques are presented. The technique has been found to be very useful to enhance various mechanical interface studies and composite behavior analyses as documented in this paper.

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